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Influence of Gas Flow Rate and Reactor Length on NO Removal Using Pulsed Power

Takao Namihira, Member, IEEE, Shunsuke Tsukamoto, Douyan Wang, Hideo Hori, Sunao Katsuki, Member, IEEE, Reuben Hackam, Fellow, IEEE, Hidenori Akiyama, Fellow, IEEE, Masaru Shimizu, and Kenichi Yokoyama

Abstract—A short duration of 100-ns pulsed power has been used to remove nitric oxide (NO) in a mixture of nitrogen, oxygen, water vapor, and NO, simulating flue gases from a power station. The effects of the gas flow rate, the reactor length, and the pulse repetition rate on the percentage of NO removal and its energy efficiency are reported. The percentage of NO removal at a fixed gas flow rate increased with increasing pulse repetition rate due to the increased energy into the discharge. At a fixed pulse rate, the removal of NO increased with decreasing gas flow rate due to the increased residence time of the gas in the discharge reactor, thus facilitating the creation of increased radicals of O and N which then decreased NO. The energy removal efficiency of NO (in mol/kWh) decreased with increasing gas flow rate and increasing removal ratio of NO. The removal of NO increased with increasing energy density (J/l) input into the discharge at different reactor length.

Index Terms—Coaxial reactor, energy efficiency, flue gases, NO removal, pulsed power.

I. INTRODUCTION

A n increasing number of power stations in the world have recently been given [3] and, therefore, they are omitted here for brevity. Only details relevant to the present experiments are briefly described here for completeness. In this work, the Blumlein line generator have recently been given [3] and, therefore, they are omitted here for brevity. Only details relevant to the present experiments are briefly described here for completeness. In this work, the Blumlein line generator had an impedance of 300 Ω and a pulse duration of 100 ns using six 10 m cables. The cable used had 0.1 nF/m and 0.25 μF/m, which gives characteristic impedance of 50 Ω. The pulse duration was defined as the full-width at half-maximum (FWHM) of the voltage waveform.

A mixture of gases was used to simulate the flue gases from a thermal power station. Gas cylinders of N₂, 0.09% NO with the balance N₂, and O₂ were used to obtain the required mixture. Water vapor was mixed with the gas mixture by passing it through water at a constant temperature. In the present work, the gas composition of the simulated exhaust gases were NO of 200 ppm (part of NO per million parts of molecules of the mixture), 5% of O₂, 2.7% of H₂O and the balance N₂ at an atmospheric pressure (1.01 × 10⁵ Pa) and 300 ± 4 K. The percentage of the pressure of water vapor in the gas mixture was controlled by maintaining a constant temperature of water [4]. The gas flow rate was varied from 2 to 12 l/min, and is reported here at an atmospheric pressure and corrected to 273 K. The gas flow rate was controlled by a mass flow controller (SEC-E440J, STEC Inc., Japan).

The reactor employed concentric coaxial cylindrical electrodes. The central rod made of stainless steel, 0.5 mm in diameter was placed concentrically in a copper cylinder having 76 mm in internal diameter. In the present work, the applied voltage from the Blumlein line generator to the reactor was measured using a resistive divider (1 Ω/10 kΩ), which was connected between the central electrode of the reactor and the ground. The positive polarity of the applied voltage was used, as it was more effective than the negative polarity [1], [6]. The discharge current in the reactor was measured using a Rogowski coil (Pearson current monitor model 2878, Pearson
Fig. 1. Typical waveforms of (a) applied voltage and (b) discharge current in the reactor for different gas flow rates. Conditions: Reactor length, 250 mm; gas pressure, $1.01 \times 10^5$ Pa; gas temperature, $300 \pm 4$ K; initial concentration, NO = 200 ppm, $O_2 = 5\%$, $H_2O = 2.7\%$ and balance $N_2$; series resistance with reactor, $0.1 \Omega$.

III. RESULTS AND DISCUSSIONS

A. Effects of Gas Flow Rate

1) Voltage and Current Wave Shapes: Fig. 1 shows typical waveforms of applied voltage (a) to and the discharge current (b) in the coaxial reactor for varying gas flow rates. The applied voltage had almost the same peak of 67.6 ± 1.4 kV and nominal pulse width of 100 ns for all gas flow rates in the range 2 to 12 l/min [Fig. 1(a)]. The negative voltage swing following the positive peak was due to impedances mismatch between the Blumlein line generator and the reactor. The peak of the current was constant at 221.9 ± 3.4 A for all gas flow rates used here [Fig. 1(b)].

2) Energy Input to the Discharge Per Pulse: Fig. 2 shows the input energy to the discharge per pulse for different gas flow rates and for durations of up to 500 ns. It should be noted that the current has decreased to 0 at about 400 ns [Fig. 1(b)]. The input energy ($\int V \times Idt$ per pulse was calculated from the voltage $V$ (in V) and current $I$ (in A) waveforms and $t$ (in s). The input energy to the reactor was constant at 0.89 ± 0.03 J/pulse for all gas flow rates in the range 2 to 12 l/min. This is because both the voltage and the current wave shapes (Fig. 1) were not affected by the large variation of the gas flow rate.

3) NO and NO$_2$ Reductions: Fig. 3 shows the final concentrations of NO (a) and NO$_2$ (b), respectively, for different gas flow rates and as a function of the pulse repetition rate after they have reached saturations. This took typically about 5 min after applying the pulsed power. It is observed from Fig. 3(a) that the concentration of NO decreased with increasing pulse repetition rate from 2 to 12 pps at a constant gas flow rate. Typically, the concentration of NO decreased from an initial value of 200 ppm to 146 ppm at 2 pps and to 38 ppm at 12 pps when the gas flow rate was 2 l/min [Fig. 3(a)]. This is because the energy input into the discharge increased with increasing the pulse repetition rate, and this resulted in an increased production of the O and N radicals, which then reduced NO via the following reactions [7]:

$$O + NO + M \rightarrow NO_2 + M \quad (1)$$

where M is a third body, which can be said N$_2$. The reaction rate of $k_1 = 6.9 \times 10^{-32}$ cm$^3$/s [8] and

$$N + NO \rightarrow N_2 + O \quad (2)$$

with a reaction rate of $k_2 = 5.9 \times 10^{-11}$ cm$^3$/s [9].

At a constant pulse repetition rate, the concentration of NO decreased with decreasing gas flow rate. The larger reduction of NO with decreasing gas flow rate (Fig. 3) is attributed to the increasing residence time of the gas in the reactor. With longer
Fig. 3. Final concentrations of (a) NO and (b) NO₂ as a function of pulse repetition rate for different gas flow rates. Nominal pulse width, 100 ns; peak voltage, 67.6 kV; peak current, 221.9 A; reactor length, 250 mm; other conditions as in Fig. 1. ● 2 l/min; ■ 4 l/min; ▲ 6 l/min; ◆ 8 l/min; ○ 10 l/min; × 12 l/min.

Fig. 4. Dependence of the energy efficiency of the removal of (a) NO and (b) NOₓ on the removal ratios of NO and NOₓ, respectively, for different gas flow rates. Conditions and symbols as in Fig. 3.

Energy Efficiency of NO Removal: Fig. 4 shows the dependence of the energy efficiencies (in mol/kWh), NOₑ of NO [Fig. 4(a)] and NOₓₑ of NOₓ [Fig. 4(b)], respectively, on the removal ratios of NO and NOₓ (≡ NO + NO₂). The removal ratio of NO (NOᵣ, in%) is given by

\[
NOᵣ = \frac{NOᵣ - NOᵣ}{NOᵣ} \times 100
\]

where NOᵣ (in ppm) and NOᵣ (in ppm) are the initial (before treatment) and the final (after treatment) concentrations of NO in the gas mixture, respectively. The removal ratio of NOₓ (NOₓᵣ, in%) is defined in a similar way to that of eq. (6). In the present work, the concentration of NOᵣ was 200 ppm. The energy efficiency of the removal of NO (NOₑ, in mol/kWh) is given by

\[
NOₑ = \frac{G \times (NOᵣ - NOᵣ) \times 60 \times [\text{min/hr}] \times 10^{-3}}{f \times E}
\]

where
- \(G\) gas flow rate [l/min];
- \(f\) pulse repetition rate [pps];
- \(E\) input energy to the reactor per pulse [J/pulse].

The energy efficiency of the removal of NOₓ (NOₓₑ, in mol/kWh) is also given by an equation similar to (7) except that NOₓᵣ and NOₓᵣ are used instead of NOᵣ and NOᵣ, respectively. The data in Fig. 4 can readily be converted to g/kWh since 0.03 kg of NO and 0.046 kg of NO₂ are equivalent to 1 mol and 0.0224 m³ at atmospheric pressure and 273 K.

Fig. 4 shows the dependence of NOₑ on NOᵣ (a) and NOₓₑ on NOₓᵣ (b) for different gas flow rates. The energy efficiency for NO and NOₓ removals decreased with increasing NO and NOₓ removal ratios at a constant gas flow rate. At a fixed removal ratio of either NO [Fig. 4(a)] or NOₓ [Fig. 4(b)], the energy efficiency for the removal of NO and NOₓ decreased with increasing gas flow rate. This is because a higher input energy is required to attain the same value of the NO removal efficiency.
ratio at a higher flow rate. This can be readily seen from Fig. 3(a) that, in order to attain a given final concentration of NO, larger pulse repetition rate and, therefore, a higher energy is required with increasing gas flow rate. The successive data in Fig. 4 (from left to right) with increasing $NOR$ and $NOX_R$ are plotted for increasing pulse repetition rate from 2 to 12 pps.

5) Energy Efficiencies of Removals of NO and NO$_X$: Fig. 5 shows the dependence of the final concentrations of NO (a) and NO$_X$ (b) on the input energy density to the gas mixture for different gas flow rates. The input energy density to the gas mixture ($E_d$, in J/l) is calculated using

$$E_d = \frac{f \times E \times 60[s/\text{min}]}{G}$$  \hspace{1cm} (8)$$

where $f$, $E$ and $G$ have been defined in (7). Fig. 5 shows that the final values of NO and NO$_X$ decreased with increasing energy density $J/l$ for all gas flow rates from 2 to 12 l/min. The dependence of the final values of NO and NO$_X$ at a fixed $J/l$ does not depend on the gas flow rate.

B. Effect of Reactor Length

1) Voltage and Current Waveforms: Fig. 6 shows typical waveforms of the applied voltage (a) and the discharge current (b) in the reactor for varying lengths at a fixed gas flow rate of 2 l/min. The dc charging voltage had to be increased with increasing the reactor length to maintain the peak pulse voltage at 53.9 $\pm$ 0.3 kV [Fig. 6(a)]. The peak of the pulse current increased from 70.4 to 208.9 A with increasing reactor length, respectively, from 200 to 800 mm [Fig. 6(b)]. The characteristics of these waveforms suggest that the impedance of the discharge decreased with increasing reactor length. This is because, progressively, a larger number of streamers were produced and, therefore, a larger current flow with increasing anode wire length in the longer reactors.

2) Energy Input/Pulse to the Discharge: Fig. 7 shows the input energy to the reactor per pulse for different reactor lengths. The input energy to the reactor increased from 0.206 to 0.632 J/pulse with increasing reactor length, respectively, from 200 to 800 mm. The coupling of a high-input energy into the discharge for longer reactor is useful as it leads to a larger reduction in
NO and NO\textsubscript{X}. The reason for the increase in the energy that can be fed into the discharge is due to the larger current, which is formed by a larger number of streamer channels with increasing reactor length (Fig. 6).

3) \textit{NO and NO\textsubscript{X} Removals:} Fig. 8 shows the final concentrations of NO (a) and NO\textsubscript{X} (b) as a function of pulse repetition rate for different reactor lengths. It will be observed from Fig. 8(a) that the concentration of NO decreased with increasing pulse repetition rate at a constant reactor length. This is because, and I as mentioned in Section III-A3, the energy input into the discharge increased with increasing pulse repetition rate. At a fixed pulse repetition rate the final concentrations of NO and NO\textsubscript{X} decreased with increasing reactor length (Fig. 8). This is due to the increased residence time of the gas in the reactor with increasing reactor length.

4) \textit{Energy Efficiency of NO Removal:} Fig. 9 shows the dependence of the energy removal efficiency as a function of the NO removal ratio [Fig. 9(a)] and the NO\textsubscript{X} removal ratio [Fig. 9(b)] for different reactor lengths. It will be observed that the energy efficiencies of both NO and NO\textsubscript{X} are not strongly dependent on the reactor length. Fig. 9 shows that the removal energy efficiencies, over a restricted range of the removal ratios of NO and NO\textsubscript{X}, when 800 mm long reactor is used, are slightly higher than for shorter reactors.

5) \textit{Dependence on Energy Density }J/l\textit{:} Fig. 10 shows the dependence of the final concentrations of NO (a) and NO\textsubscript{X} (b) on the input energy density to the gas mixture for different reactor lengths. Fig. 10 shows that the final concentrations of NO and NO\textsubscript{X} decrease with increasing energy density.
NOX decreased with increasing the energy density input to the discharge. At a fixed input energy density to the gas mixture, the final concentrations of NO and NOX slightly decreased with increasing reactor length in the range 200 to 800 mm. Fig. 10 shows that, in order to obtain a constant value of reduced NO and NOX, the amount of J/l required is not strongly dependent on the reactor length. This can be explained by a reference to Fig. 8, where it can be seen that a lower pulse repetition rate can be used to attain fixed values of the reduced NO and NOX. However, at the same peak pulse voltage of 53.9 kV, much larger discharge currents were present [Fig. 6(b)] for longer reactors.

IV. CONCLUSION

Pulsed power has been used to remove NOX in a mixture of N2, O2, and H2O. The following conclusions have been deduced.

1) For a fixed length of the reactor, the peak of the pulse voltage and the peak of the pulse discharge current were independent of the gas flow rate in the range 2–12 l/min when a constant charging voltage was used.
2) The energy input/pulse into the discharge was independent of the gas flow rate at a fixed reactor length.
3) The final concentrations of NO and NOX decreased with increasing pulse rate at a fixed reactor length and a fixed gas flow rate.
4) The final concentrations of NO and NOX decreased with decreasing gas flow rate at a fixed reactor length and a fixed pulse repetition rate.
5) The energy efficiency removal (mol/kWh) of NO and NOX decreased with increasing their removal ratios at a constant gas flow rate.
6) The energy efficiency removal (mol/kWh) of NO and NOX decreased with increasing gas flow rate at constant removal ratios of NO and NOX, respectively.
7) The final concentrations of NO and NOX decreased with increasing energy density (J/l) input to the discharge and were independent of the gas flow rate at a constant J/l.
8) At a fixed gas flow rate, the peak of the pulsed voltage was maintained constant by increasing the charging voltage, and the peak of the pulsed current increased, with increasing reactor length.
9) The energy input/pulse into the discharge increased with increasing reactor length.
10) At a fixed gas flow rate and a fixed pulse repetition rate, the final concentrations of NO and NOX decreased with increasing reactor length.
11) The NO and NOX energy efficiency removal at a fixed removal ratio was not strongly dependent on the length of the reactor.
12) At a fixed energy density input (J/l) the final concentrations of NO and NOX were independent of the reactor length.

REFERENCES


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